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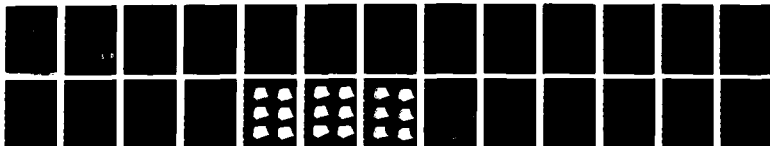
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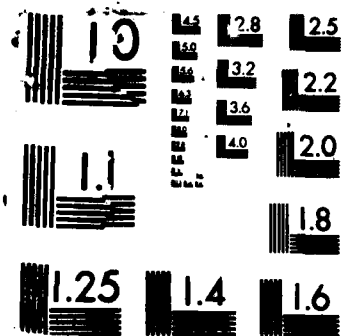
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Theory of Laser-Stimulated Processes. III. Desorption Through
Vibrational Excitation by an IR Laser

by

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in

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Theory of Laser-Stimulated Surface Processes. III. Desorption Through
Vibrational Excitation by an IR Laser.

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Abstract

Desorption of an adatom from a solid surface through vibrational excitation by an IR laser is studied using a generalized master equation approach and the isomnesic approximation. A sharp transition from no desorption to almost instantaneous desorption is seen as the bound-to-continuum population transfer rate is varied. The critical rate increases linearly with laser power.

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I. Introduction

In the field of laser-stimulated surface processes (LSSP), desorption is the most commonly studied phenomenon. As demonstrated in a number of experiments,¹⁻³ the desorption cross section can be measured unequivocally, albeit with some difficulty. One aspect of these experiments which is especially significant is the strong dependence of the desorption signal on the laser frequency. The control that this selectivity gives us in directing the course of chemical reactions on a solid surface holds the promise of establishing a totally new kind of chemistry. However, not only is there a shortage of well-characterized laser-desorption experiments, but also a clear understanding of the detailed mechanisms responsible for the selectivity has been lacking. The reader is referred to recent reviews for details.³⁻⁵

The aim of our early work on LSSP⁴⁻⁷ was to delineate the coarse features of the phenomena involved. Using models in which the adspecies (a molecule) vibrated in the attractive region of a surface potential, the degree of selectivity was shown to depend on the anharmonicity of the potential and the mismatch between the laser frequency ω_L and the fundamental vibrational frequency ω_A of the adspecies in the adsorption potential. In addition, the existence of energy feedback was made plausible.⁶ Thus, assuming that the laser photons were absorbed directly by a so-called internal active mode A of the molecule, which, in turn, was coupled to sink modes B (other modes of the molecule and phonon modes of the solid), energy was shown to return from B to A on a short time scale for specific ranges of laser power and system parameters. While this could be rationalized in terms of a slow energy dissipation rate inside the solid, leading to local buildup in the vicinity of the adsorbed molecule, it was not possible to distinguish specific mechanisms within that phenomenological framework.

More mechanistic approaches have subsequently been adopted, generally based on Morse potentials for the adspecies.⁸ Mechanisms for desorption based on pumping by internal mode excitation of the adsorbed molecule and tunnelling between degenerate bound and continuum levels of the adspecies¹⁰ have explained some features of the experimental results. In an attempt to isolate the conditions for, and onset of, selectivity, we have presented a time-dependent approach. Phenomenological time-independent coupling constants are replaced by time-dependent memory kernels, and time averages are supplanted by temporally varying distribution functions. So far, energy transfer has been studied in the absence of a desorption channel, but selective absorption has been confirmed for vanishing mismatch $\omega_L - \omega_A$. In these first-principles quantum mechanical treatments, ω_A is represented by a transition frequency $\omega_{SS'}$ (where S and S' are vibrational quantum numbers of stationary states of the adsorption potential) closest to ω_L . Furthermore, the time-dependent profile of the average vibrational energy of the adsorptive bond (the adbond) shows an initial strong increase followed by a pronounced minimum and, subsequently, another increase.¹¹ The decrease leading to the minimum is due to energy transfer to the bulk phonons; and the subsequent increase is due to feedback from the phonons to the adbond. In this paper, desorption is considered in the form of a transition from an excited bound state of the adsorption potential to the continuum. The resulting formalism is found to depart insignificantly from that of our previous work¹¹⁻¹³ in terms of interpretive power. Without recourse to massive computer calculations, it is possible to obtain time-dependent probability and energy profiles within the isomnesic approximation. Details of the desorption formalism are presented in Section III, which follows a brief summary of the time-dependent theory of vibrational energy transfer between an adatom, a solid and an IR laser in Section II. Results are presented and discussed in Section IV.

II. Theory of Vibrational Energy Transfer Between a Laser, an Adsorbate and Bulk Phonons

The essential elements of this theory¹¹⁻¹³ are: (i) a specific partitioning of the Hamiltonian of the system consisting of atoms adsorbed on a solid surface in the presence of radiation from an IR laser; (ii) generation of a closed form for the time-dependent memory kernels $K_{SS'}^{(p)}(t)$ and $K_{SS'}^{(r)}(t)$ representing the dynamical contributions of the adbond-phonon and adbond-radiation interactions, respectively; (iii) solution of Liouville's equation using the projection operator technique, which leads to the generalized master equation (GME)

$$\dot{P}_S(t) = \sum_{S' \neq S} \int_0^t dt' [K_{SS'}(t-t') P_{S'}(t') - K_{S'S}(t-t') P_S(t')] \quad , \quad (1)$$

where $P_S(t)$ is the probability of finding the adbond in the state $|S\rangle$ of an effective potential, and

$$K_{SS'}(t) = K_{SS'}^{(p)}(t) + K_{SS'}^{(r)}(t) \quad ; \quad (2)$$

and finally (iv) approximating $K_{SS'}^{(p)}(t)$ as a delta function and $K_{SS'}^{(r)}(t)$ as a constant to give the isomnesic approximation, allowing closed-form solutions of the GME.

The special properties of laser radiation are emphasized by taking a three-level system, $|S\rangle = |0\rangle, |1\rangle, |3\rangle$, one of whose transitions is resonant with the laser, and another equal to the Debye frequency for the phonons (Fig. 1). For this case, application of the Laplace transform method to the GME leads to a third-order polynomial equation.^{12,13} In the absence of desorption, the total bound-state probability is seen to be conserved, and a steady state is eventually achieved.

111. Desorption

Desorption corresponds to transfer of population from the manifold of bound levels of the adsorption potential to the continuum. We introduce a phenomenological constant transition rate R from the highest bound level $|\sigma\rangle$ attainable through the combination of resonant laser and phonon excitations, to continuum states $|\kappa\rangle$. In our case, $|\sigma\rangle = |3\rangle$. As a result we must include an extra term $-RP_\sigma(t)$ in the GME for $\dot{P}_\sigma(t)$, so that

$$\dot{P}_S(t) = \sum_{S' \neq S} \int_0^t dt' [K_{SS'}(t-t') P_{S'}(t') - K_{S'S}(t-t') P_S(t')] , \quad S \neq \sigma \quad (3)$$

and

$$\dot{P}_\sigma(t) = \sum_{S' \neq \sigma} \int_0^t dt' [K_{\sigma S'}(t-t') P_{S'}(t') - K_{S'\sigma}(t-t') P_\sigma(t')] - RP_\sigma(t) . \quad (4)$$

In the isomnesic approximation, we replace Eq.(2) by

$$K_{SS'}(t) = \Omega_{SS'} \delta(t) + k_{SS'} , \quad (5)$$

where $\Omega_{SS'}$ and $k_{SS'}$ are constants in time. We can write, for all S ,

$$\begin{aligned} \dot{P}_S(t) = & \sum_{S' \neq S} \{ k_{SS'} \int_0^t dt' P_{S'}(t') - k_{S'S} \int_0^t dt' P_S(t') \\ & + \Omega_{SS'} P_{S'}(t') - [\Omega_{S'S} + R\delta_{S\sigma}] P_S(t) \} . \end{aligned} \quad (6)$$

Differentiating and taking the Laplace transform, we get

$$\begin{aligned} s^2 P_S(s) - sP_S(0) - \dot{P}_S(0) = & \sum_{S' \neq S} \{ k_{SS'} \rho_{S'}(s) - k_{SS'} P_S(s) \\ & + \Omega_{SS'} [s\rho_{S'}(s) - P_{S'}(0)] \\ & - [\Omega_{S'S} + R\delta_{S\sigma}] (s\rho_S(s) - P_S(0)) \} \end{aligned} \quad (7)$$

$$= \sum_{\text{all } S} \{ k_{SS'} \rho_{S'}(s) + \Omega_{SS'} (s\rho_{S'}(s) - P_{S'}(0)) \} , \quad (8)$$

where we have defined "diagonal" elements of the matrices \underline{k} and $\underline{\Omega}$ as

$$k_{SS} = - \sum_{S' \neq S} k_{S'S} , \quad (9)$$

$$\Omega_{SS} = - \sum_{S' \neq S} \Omega_{S'S} - R \delta_{SS} \quad (10)$$

and $\mathcal{P}_S(s)$ is the Laplace transform of $P_S(t)$.

For the previously studied case of three bound levels, we get

$$\underline{\Omega} = \begin{pmatrix} -A & B & 0 \\ A & -B & 0 \\ 0 & 0 & -R \end{pmatrix} \quad (11)$$

where,

$$A = \Omega_0 \alpha \quad (12)$$

$$B = \Omega_0 (1 + \alpha) \quad (13)$$

$$\alpha = C'/2\chi \quad (14)$$

$$\chi = \hbar \omega_D / 2k_B T \quad (15)$$

$$C' = e^{-p\chi} \quad (16)$$

p being a fitting parameter involved in the integration over the phonon density of states, k_B the Boltzmann constant, and T the temperature. The local rate Ω_0 due to the phonon-adsorb interaction is obtained from $\Omega_{SS'}$:

$$\Omega_{SS'} = \Omega_0 (1/2) [1 - (\omega_{SS'}/|\omega_{SS'}|) + C'/\chi] \delta_{|\omega_{SS'}|, \omega_D} \quad (17)$$

Inversion of the Laplace transform equation involves the solution of the polynomial equation

$$q(s) = \det \underline{M} = 0 \quad (18)$$

where

$$\underline{M} = \underline{k} + s\underline{\Omega} - s^2 \underline{A} \quad (19)$$

\underline{A} being the unit matrix. Thus we need the roots of the polynomial equation

$$\begin{vmatrix} -(s^2 + As) & Bs & 0 \\ As & -(s^2 + Bs + k_0) & k_0 \\ 0 & k_0 & -(s^2 + Rs + k_0) \end{vmatrix} = 0 \quad (20)$$

where $k_0 = k_{SS}$, for the pair of levels resonant with the laser. Equation (20) is a quartic for $R \neq 0$ and a cubic for $R = 0$. The roots can, in principle, be obtained in closed form, but, because of their complicated nature, are of little practical use in our analysis. We have instead adopted a numerical approach whose details we shall omit. The resulting probability profiles are used to define the desorption probability P_D and average adbond energy as follows:

$$P_D(t) = P_T(0) - \sum_S P_S(t) , \quad (21)$$

$$E(t) = \sum_S \epsilon_S P_S(t) , \quad (22)$$

where ϵ_S is the energy eigenvalue corresponding to $|S\rangle$, and

$$P_T(0) = \sum_S P_S(0) . \quad (23)$$

The value of $P_T(0)$ is slightly smaller than unity because the set of levels $\{S\}$ used in the GME does not include all bound states of the effective adbond potential.

The specific nature of the parameters A , B and k_0 is of interest from an analytic point of view. Their form can be derived, from exact expressions for the memory kernels k_{SS} , presented in earlier work,⁸ by using the IA, Eq. (5). In light of Eqs. (12)-(16), the parameters A , B and k_0 involve Ω_0 and k_{SS} , which are given by

$$\Omega_0 = A_p B_p \chi^2(\pi/2\omega_D) \epsilon_{SS}, \quad (\text{for } |\omega_{SS}| = \omega_D) \quad (24)$$

and

$$k_{SS} = A_r (z_{SS})^2 \delta_{|\omega_{SS}|, \omega_L} , \quad (25)$$

where

$$A_p = 8(D_e^{(0)}/\hbar)^2, \quad (26)$$

$$B_p = 6(\beta^{(0)}k_B T)^2/M_s \hbar \omega_D^3, \quad (27)$$

$$\Xi_{SS'} = \int_{\mu\mu'} (-1)^{\mu\mu'} (e^{\overline{u}^2/2})^{\mu^2+\mu'^2} E_{S'S}^{(\mu)} E_{SS'}^{(\mu')}, \quad (28)$$

$$E_{SS'}^{(\mu)} = \langle S | e^{-\mu(z-z_0^{(0)})} | S' \rangle, \quad (29)$$

$$A_r = (I e_0^2 / \epsilon_0 c \hbar^2 \beta^{(0)2}), \quad (30)$$

$$z_{SS'} = \langle S | z | S' \rangle. \quad (31)$$

The quantities in Eqs. (26)-(31) involve the following:

$D_e^{(0)}$, $\beta^{(0)}$: well depth and steepness parameters for an effective Morse potential between the surface atom and the adatom

M_s : atomic mass of the solid

μ, μ' : take on the values 1 and 2

\overline{u}^2 : mean square displacement of the atoms of the solid from equilibrium.

z : position normal to the solid surface measured with respect to the equilibrium position of the outer most lattice atom.

$z_0^{(0)}$: location of the minimum of the effective Morse potential between the surface atom and the adatom

I : laser intensity

e_0 : charge difference between the adspecies and the surface

ϵ_0 : permittivity of free space.

The strengths of the adbond-solid interaction and the laser-adsorbate interaction are represented by Ω_0 and k_0 , respectively. It has been demonstrated that the two must be comparable in order for the system to manifest significant synergistic, selective effects.

IV. Results and Discussion

With the inclusion of a unidirectional desorption channel, we expect the total population of bound states to be depleted monotonically, i.e., $P_D(t)$ must grow monotonically to a maximum possible value of $P_T(0)$. Similarly, since desorption is due to transfer of population from the high-lying level $|o\rangle$ to the continuum, the average adbond energy $\mathcal{E}(t)$ might be expected to increase with time. However, because of the competition between laser pumping, which populates the level $|o\rangle$, and desorption, which depletes it, the final behavior of $\mathcal{E}(t)$ is not as simple as that of $P_D(t)$. For vanishing R , $\mathcal{E}(t)$ should have the familiar nondesorptive profile.⁸⁻¹⁰ For values of R comparable to the effective laser pumping rate (characterized by, but not equivalent to, k_0) the behavior of $\mathcal{E}(t)$ cannot be predicted on intuitive grounds. For the other extreme, namely for laser pumping rates much smaller than the rate R , the average adbond energy is expected to rise quickly as population is depleted uniformly from the lower-lying levels and does not return to them. Eventually the system should be completely desorbed and $\mathcal{E}(t)$ should become zero, implying the complete absence of populated bound states.

Our results for the prototype system of references 11-13 bear out these expectations. Figures 2 through 10 show the desorption probabilities $P_D(t)$ and the associated average adbond energies $\mathcal{E}(t)$ for laser powers ranging from 10 W/cm^2 to 10^9 W/cm^2 and for desorption rates spanning five orders of magnitude. Notice that the time scales over which the system exhibits

significant dynamical effects increase with decreasing laser power, as expected. Significant dynamical changes take place more slowly for lower laser powers.

Both $P_D(t)$ and $\mathcal{E}(t)$ display a transitional dynamical state of the system for a certain narrow range of desorption rates. For rates below this, the system is relatively tranquil, with the laser causing almost no change in the adsorbed system. The desorption probability changes imperceptibly over quite a large time period, and $\mathcal{E}(t)$ oscillates about an eventual steady-state value. The behavior of the system for R larger than this transitional range is radically different and violent. The adbond energy $\mathcal{E}(t)$ and the desorption probability $P_D(t)$ increase rapidly and monotonically to new equilibrium values, the rise time decreasing rapidly with increasing R . The changes in behavior of the system with I and R are quantified in Fig. 11, where regions of oscillatory (O), nonoscillatory (N) and transitional (T) behavior are demarcated. For the range of R studied, namely $10^{-6} \text{ ps}^{-1} \leq R \leq 1 \text{ ps}^{-1}$, both the cessation of slow oscillatory behavior and the onset of fast sigmoid behavior takes place at higher laser powers for larger transition strengths R . While the actual locations of the boundaries between the regions O and T and the regions T and N (obtained by inspection from Figs. 2-10) are crude estimates, the width of region T is uniform and the edges essentially linear. The trends depicted in Fig. 11 are important from the point of view of selective laser photochemistry. Thus, for a given value of R , laser powers in the region O will cause a very slow accumulation of vibrational energy in the adbond, while laser powers in regions T and N will excite the adbond very quickly and cause almost instantaneous desorption.

The actual experimental situation is complicated by the fact that, in general, there is a contribution to R due to the phonon field itself. The strong interaction between the adbond and phonons would therefore dominate the

desorption dynamics, and sampling a range of R to test for the two types of behavior would be difficult. For the case of a bound state $|\beta\rangle$ embedded in the continuum, however, resonant excitation from $|\alpha\rangle$ to $|\beta\rangle$ using another laser provides a practical means of varying R . A fairly sharp increase in the desorption cross section as a function of laser intensity would be the signature of the transition from a mostly bound to a mostly desorbed state. The whole process is strongly bond-selective, and for the case of a variety of adsorbed species, different pairs of lasers should cause different adspecies to desorb. Also, for low-power operation with lasers whose frequencies do not match system transitions, no sudden transition would be observed.

Purely thermal effects can be expected to dominate for the case of significant departure from the two generic resonance conditions shown in Fig. 1. These conditions are designed to highlight the mechanism responsible for selective effects associated with laser-stimulated desorption. An additional factor that exerts a strong influence on the dynamics of LSSP in general is the rate of dissipation of vibrational energy from the Debye phonons to the rest of the allowed phonon states. The resonance condition involving the Debye frequency translates to an energy transfer rate between the adbond and the solid which is much larger than the phonon dissipation rate, a factor of central importance for the results of the model presented here. The implied 'localization' of vibrational energy in a thin slice of the phonon density of states is the k -space analog of spatial localization of energy, but with additional many-body effects exhibited as oscillations of $\xi(t)$. Thus, for example, if the phonon energy dissipation rate is comparable to or greater than the laser pumping rate, the 'feed' level $|\beta\rangle$ for desorption will not be sufficiently populated to exhibit the large increase in average adbond energy due to selective resonant excitation. On the other hand, if the phonon energy dissipation rate is comparable to or greater than the energy transfer rate between the adbond and the Debye phonons, the

feedback effects represented by the oscillations of $\xi(t)$ would not be evident on the time scales of Figs. 2-7. The occupation of Debye phonons would not get sufficiently large for the back transfer of vibrational energy from the solid to the adbond to be significant. However, within the framework of the assumed resonance conditions, the many-body effects lead to the very interesting phenomena described earlier. Experimentally, the sharpness of the resonances will, of course, determine if the selective effects are observable directly, or if they will be obscured by nonresonant thermal effects. It would be interesting to investigate the possibility of separating the selective and the nonselective effects in the experimental results. Knowledge of the behavior of P_D or ξ as a function of R for a fixed time (perhaps an asymptotic limit) due to purely thermal effects is necessary for such a separation.

The GME for the desorptive case, Eq. (6), suggests a similarity between the treatment of the phonon-adbond interaction, i.e., a delta function limit for $K_{SS}^{(p)}(t)$, and that of the desorptive term. While there are two continua present - the phonon distribution on the one hand and the desorption states on the other - it is not clear that the two can lead to similar memory effects. If the $|e\rangle$ -to-continuum transition is primarily to a very small slice, it may be necessary to treat this channel analogously to the laser-adbond interaction, namely with an isomnesic kernel. A detailed treatment with well-defined excitation mechanisms is necessary for a better understanding of the role of the desorption channel.

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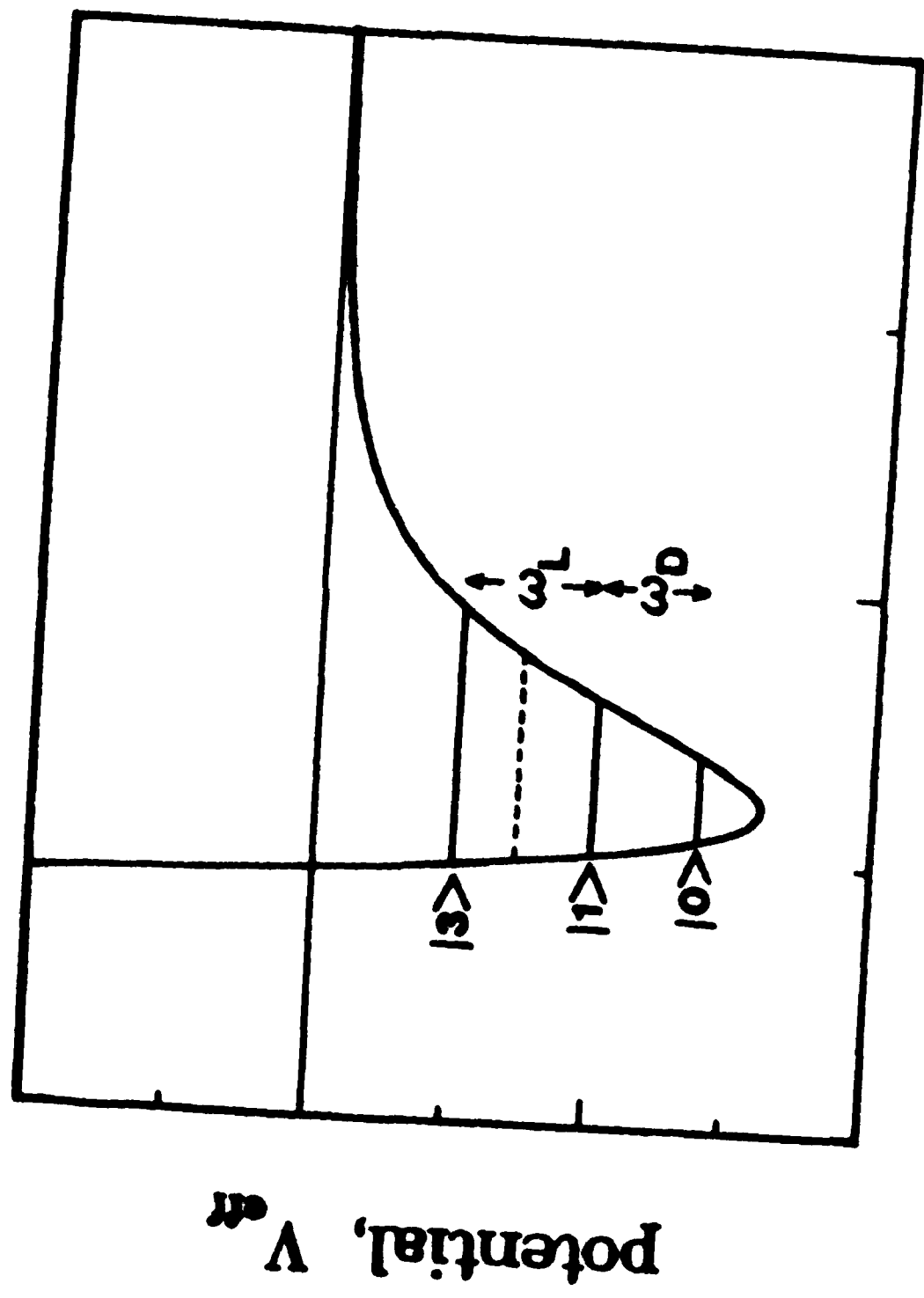
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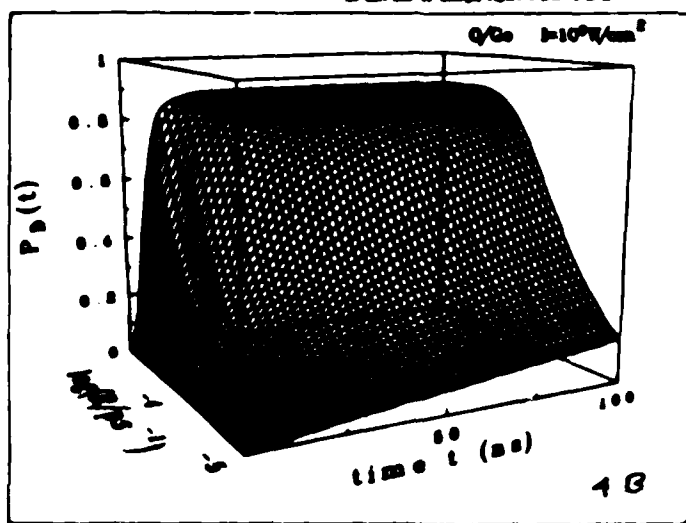
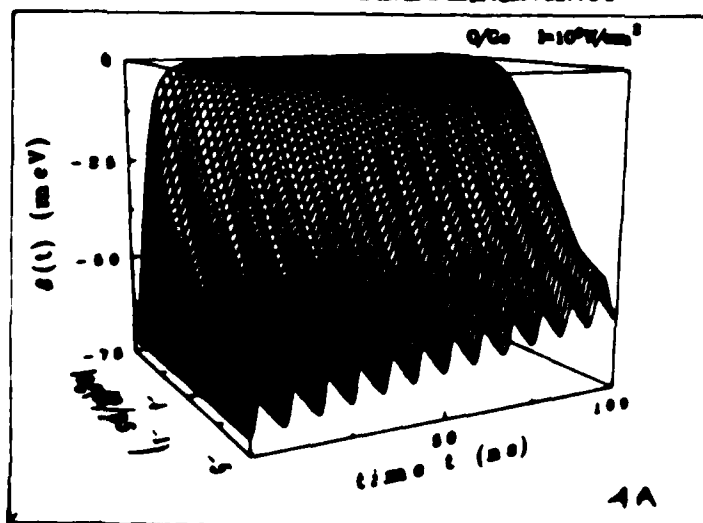
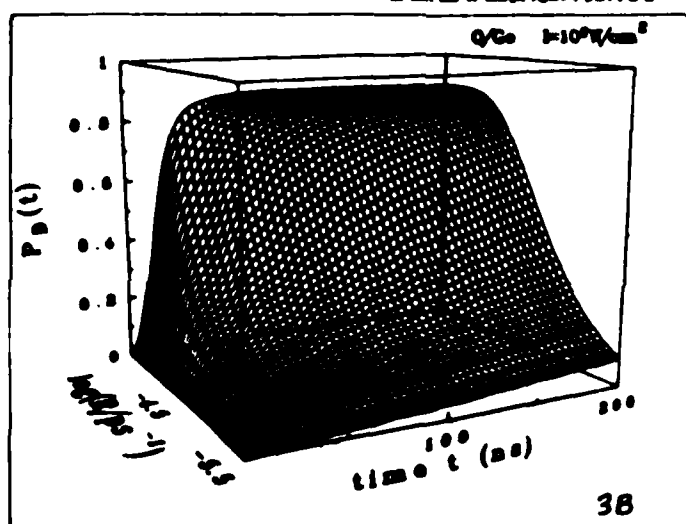
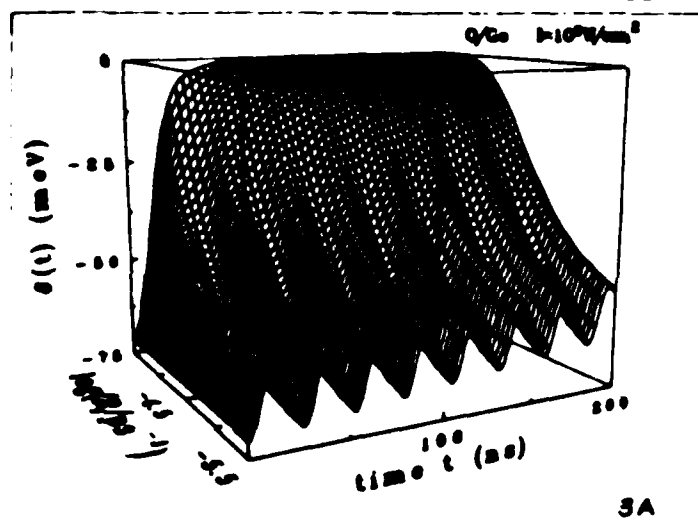
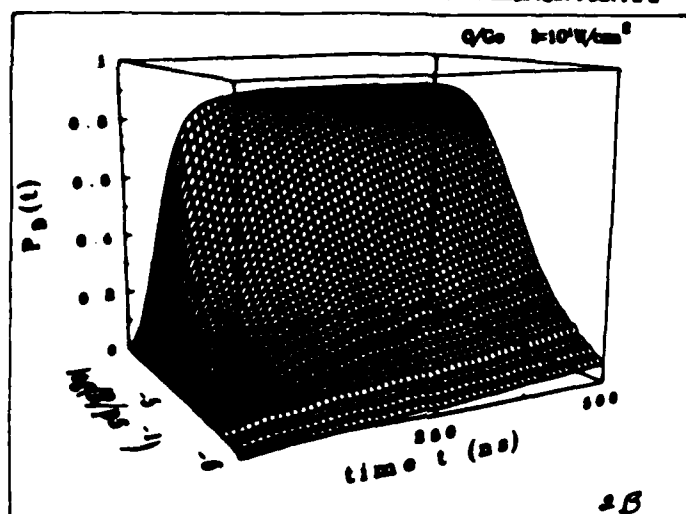
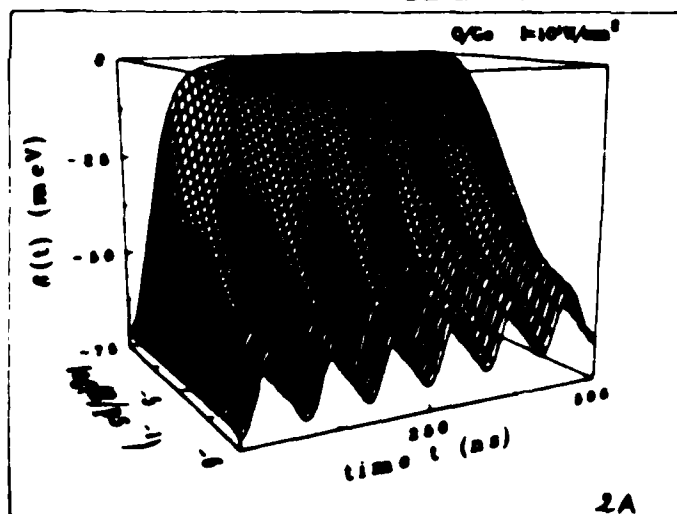
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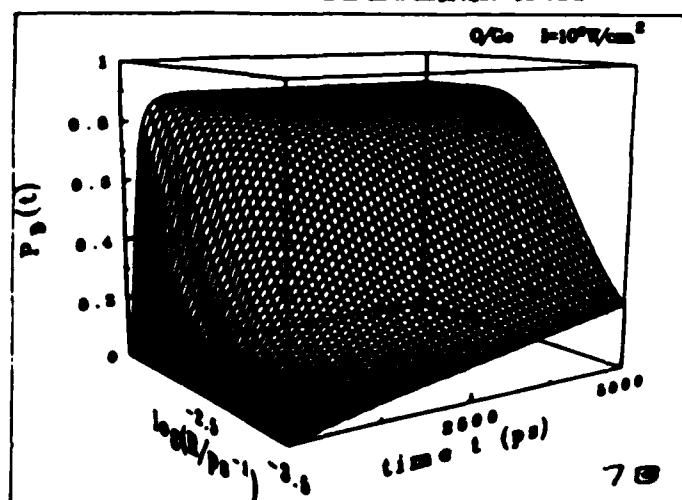
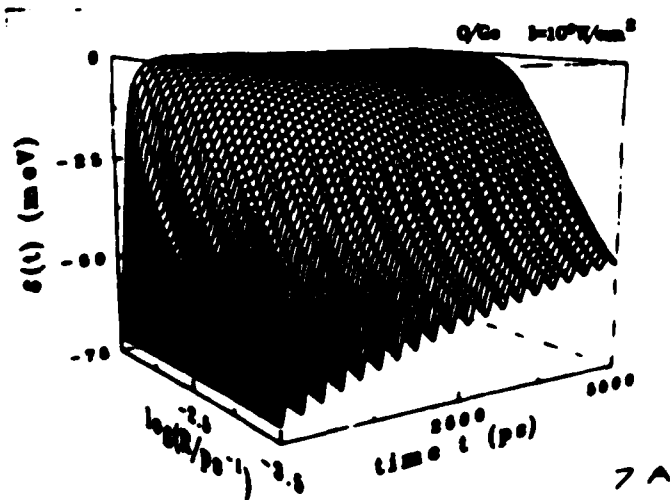
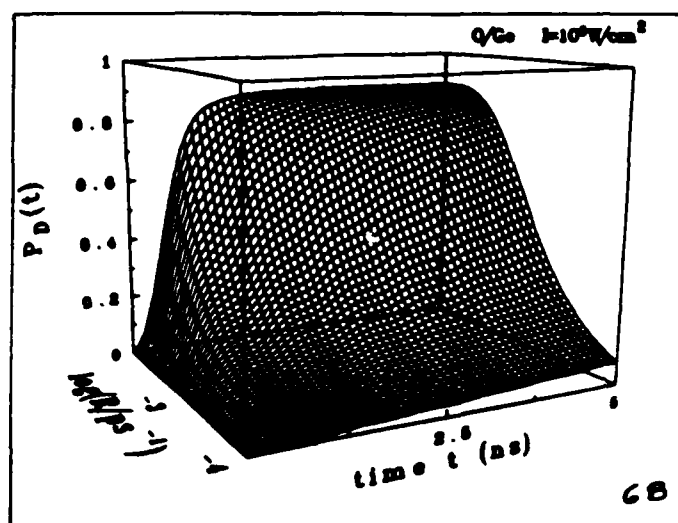
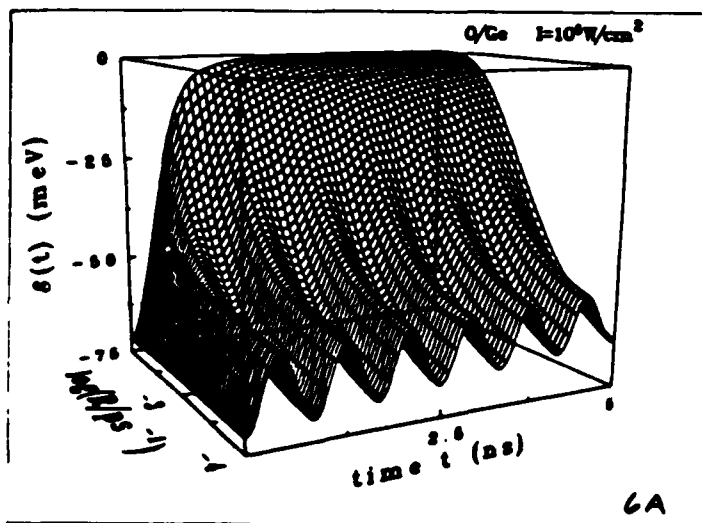
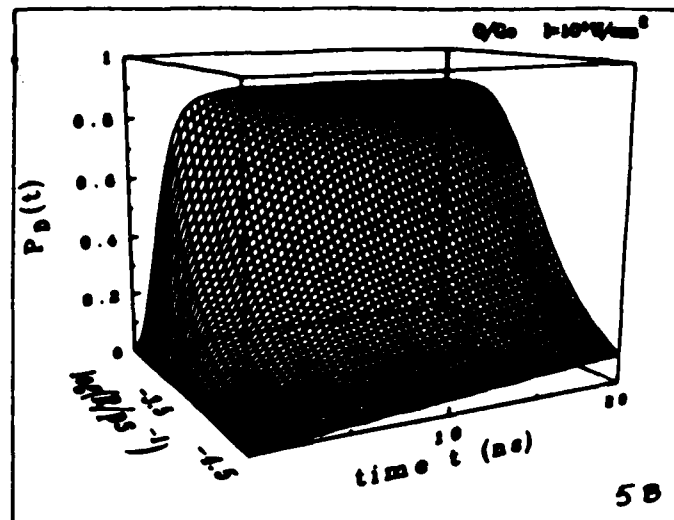
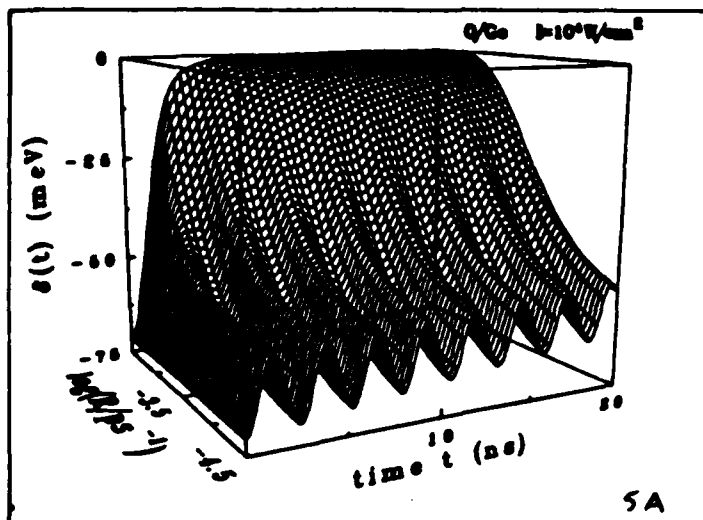
- Fig. 1. Choice of bound vibrational states of the effective potential $V_{\text{eff}}(z)$ between an adatom and a solid.
- Fig. 2. (A) Average adbond energy $\bar{E}(t)$ and (B) Desorption probability $P_D(t)$ of the system in Fig. 1 for a range of values of the transition strength R and laser power $I = 10 \text{ W/cm}^2$.
- Fig. 3. As in Fig. 2 for $I = 10^2 \text{ W/cm}^2$.
- Fig. 4. As in Fig. 2 for $I = 10^3 \text{ W/cm}^2$.
- Fig. 5. As in Fig. 2 for $I = 10^4 \text{ W/cm}^2$.
- Fig. 6. As in Fig. 2 for $I = 10^5 \text{ W/cm}^2$.
- Fig. 7. As in Fig. 2 for $I = 10^6 \text{ W/cm}^2$.
- Fig. 8. As in Fig. 2 for $I = 10^7 \text{ W/cm}^2$.
- Fig. 9. As in Fig. 2 for $I = 10^8 \text{ W/cm}^2$.
- Fig. 10. As in Fig. 2 for $I = 10^9 \text{ W/cm}^2$.
- Fig. 11. Regions of oscillatory (O), and nonoscillatory (N) behavior of the system of Fig. 1 for $10^{-6} \text{ ps}^{-1} \leq R \leq 1 \text{ ps}^{-1}$ and $10 \text{ W/cm}^2 \leq I \leq 10^9 \text{ W/cm}^2$, and the transitional region (T) between the two.

AO Bart and TP George, Figure 1

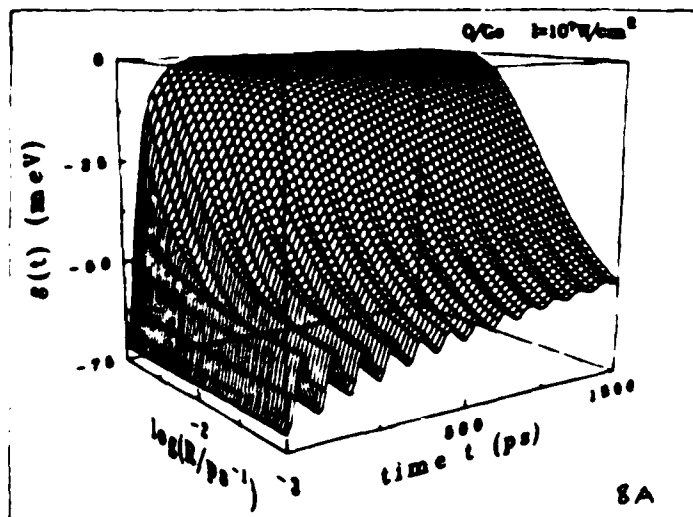


distance from surface, z

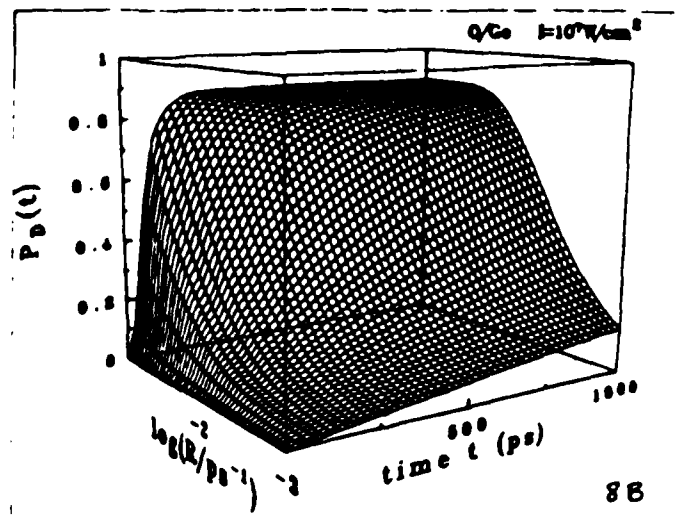




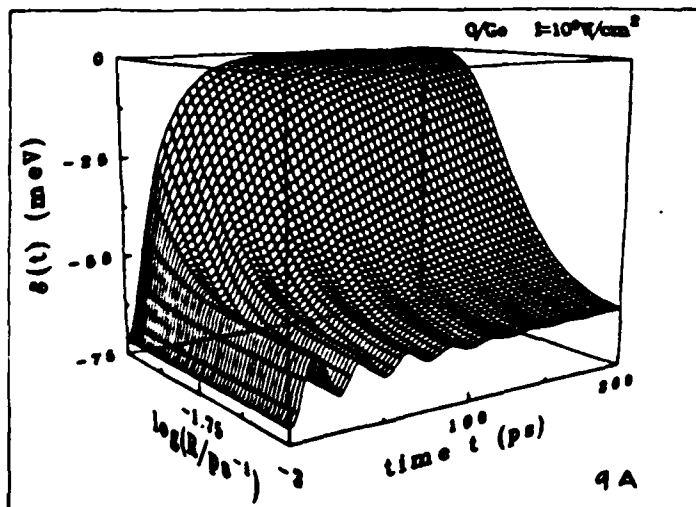
8A and 8B, Figure 8, Sheet 1 of 2



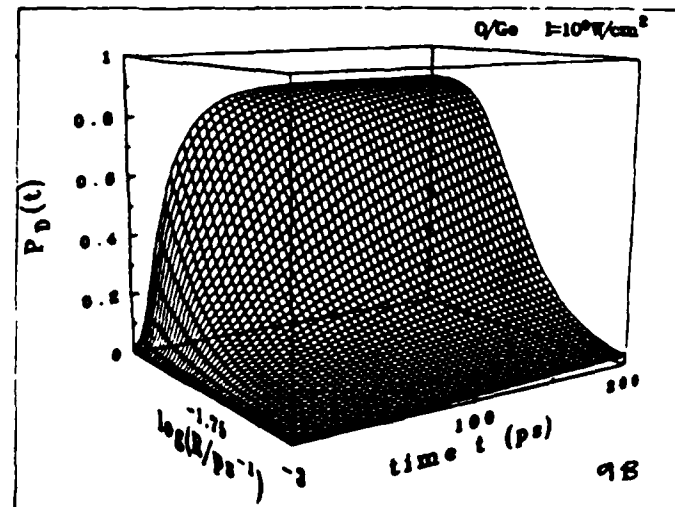
8A and 8B, Figure 8, Sheet 2 of 2



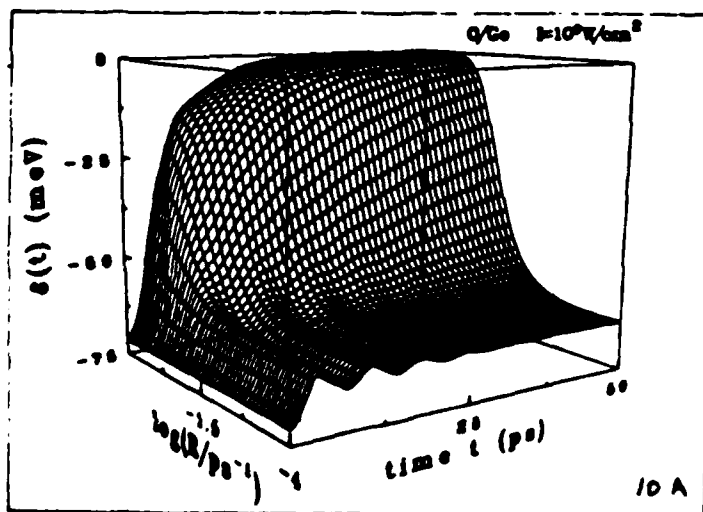
9A and 9B, Figure 9, Sheet 1 of 2



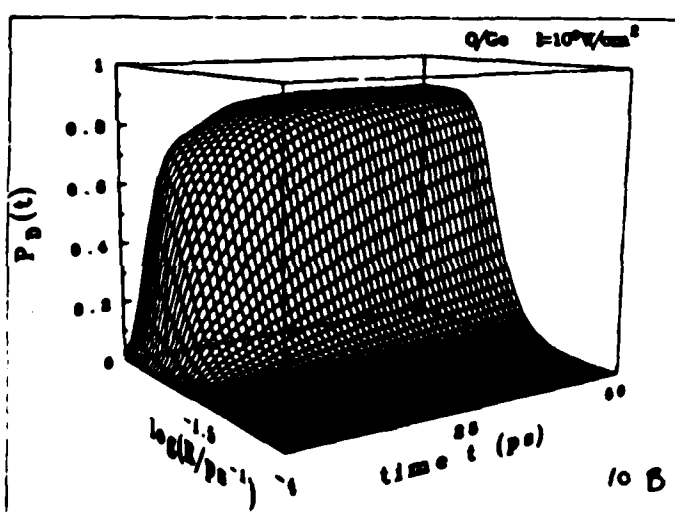
9A and 9B, Figure 9, Sheet 2 of 2

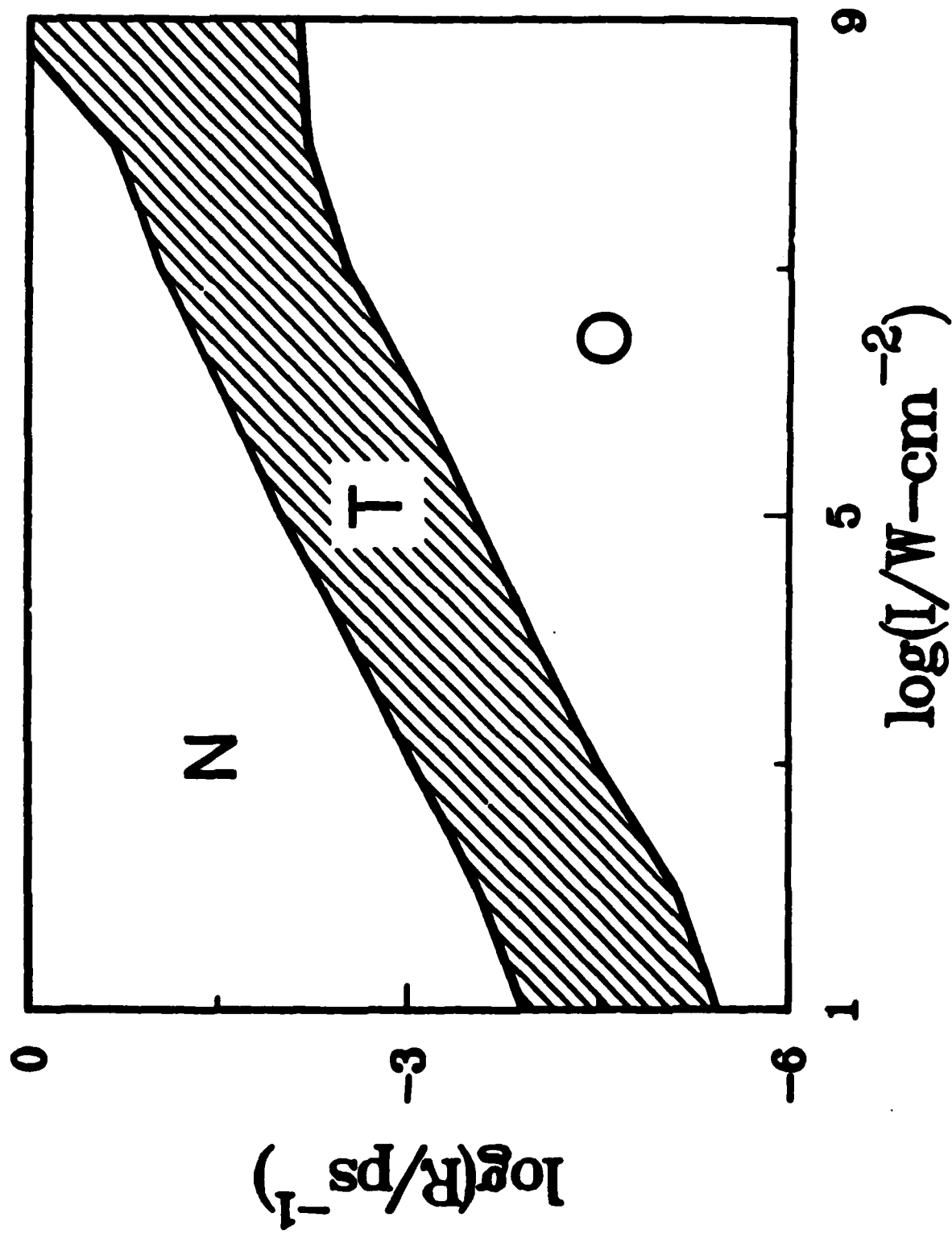


10A and 10B, Figure 10, Sheet 1 of 2



10A and 10B, Figure 10, Sheet 2 of 2





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